

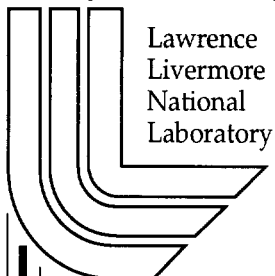
# What is a shock wave to an explosive molecule?

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# WHAT IS A SHOCK WAVE TO AN EXPLOSIVE MOLECULE?

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An explosive molecule is a metastable chemical species that reacts exothermically given the correct stimulus. Impacting an explosive with a shock wave is a "wake-up call" or "trigger" which compresses and heats the molecule. The energy deposited by the shock wave must be distributed to the vibrational modes of the explosive molecule before chemical reaction can occur. If the shock pressure and temperature are high enough and last long enough, exothermic chemical decomposition can lead to the formation of a detonation wave. For gaseous, liquid, and perfect single crystal solid explosives, after an induction time, chemical reaction begins at or near the rear boundary of the charge. This induction time can be calculated by high pressure, high temperature transition state theory. A "superdetonation" wave travels through the preshocked explosive until it overtakes the initial shock wave and then slows to the steady state Chapman-Jouguet (C-J) velocity. In heterogeneous solid explosives, initiation of reaction occurs at "hot spots" created by shock compression. If there is a sufficient number of large and hot enough "hot spots," these ignition sites grow creating a pressure pulse that overtakes the leading shock front causing detonation. Since the chemical energy is released well behind the leading shock front of a detonation wave, a mechanism is required for this energy to reinforce the leading shock front and maintain its overall constant velocity. This mechanism is the amplification of pressure wavelets in the reaction zone by the process of de-excitation of the initially highly vibrationally excited reaction product molecules. This process leads to the development of the three-dimensional structure of detonation waves observed for all explosives. In a detonation wave, the leading shock wave front becomes a "burden" for the explosive molecule to sustain by its chemical energy release.

## INTRODUCTION

What is a shock wave to an explosive molecule? There are several answers to this question depending upon the strength and time duration of the shock pulse. Since an explosive molecule (or a mixture of fuel and oxidizer molecules) is inherently metastable, it requires only an increase in its internal energy to overcome its activation energy barrier to reaction. This decomposition process may eventually become highly exothermic and cause deflagration (subsonic reaction) or detonation (supersonic reaction). So a shock wave is the "wake-up call" or the "trigger" that causes the molecule to release its chemical energy. The shock pulse must be of sufficient strength and time duration or self-sustaining exothermic chemical reaction does not occur. In a heterogeneous solid explosive, a weak shock wave can create a compressed material that does not react when

subjected to subsequent shock waves. Strong shock waves create reactive flows in their wake. These reactive flows can couple to, reinforce and strengthen the shock front. The result is a detonation wave, in which the leading shock wave front is sustained by the chemical energy released behind it. Then the shock front is not only a "trigger" but also a "burden" to the explosive molecule since it must be sustained by its exothermic chemical reaction.

Therefore a shock wave can be many different things to an explosive molecule. In this paper the current state of knowledge and future research directions for each of these regimes are briefly discussed in order of increasing pressure.

## NON-SHOCK IMPACT IGNITION

When a heterogeneous solid explosive charge is subjected to a low velocity impact that produces a

few kilobars of pressure, a two-stage compression wave is formed. This wave consists of an elastic wave that propagates through the explosive at longitudinal sound velocity followed by a plastic wave traveling at lower velocity (1,2). Within the flow field produced by the plastic wave, regions of the explosive can be heated by void collapse, friction, shear, and other possible mechanisms (3). "Hot spots" are formed and can ignite and grow into an explosive energy release. Most of these ignitions result in subsonic deflagration waves driven by heat transfer from the hot reaction products into the surrounding explosive molecules. Impact ignition is one of the most important explosive safety concerns, because it is caused by the smallest amount of energy delivered to the explosive molecules. Several tests have been developed to study impact: drop hammers; drop weight impact machines; Skid tests; the Susan test; etc. In recent years, the Steven Test at LLNL (4) and its modified version at LANL (5) have been used to yield quantitative experimental data that can be simulated with reactive flow computer models. The Steven Test and other impact studies have resulted in an improved understanding of impact ignition.

### WEAK SHOCK COMPRESSION

At slightly higher pressures, the elastic and plastic waves merge into a relatively weak shock wave (1,2). For homogeneous explosives, these shocks compress and heat the explosive molecules slightly, but little or no chemical decomposition occurs. For some heterogeneous solid explosives, there exists a narrow range of shock pressures in which all of the voids and other inhomogeneities can be compressed without creating growing hot spot reactions. The resulting fully dense explosive material can not be shock initiated by subsequent strong shock waves or even detonation waves. This phenomenon is called "dead pressing" or "shock desensitization"(6). Depending on the application, this can be a useful or a frustrating property of explosive molecules.

### HOMOGENEOUS EXPLOSIVES

Homogeneous explosives include gases, liquids without bubbles or suspended solids, and perfect crystals of solid explosives. In these materials, planar shock waves uniformly compress and heat the explosive molecules. There has long been some debate about the definition of the thickness of a

shock wave. Zeldovich and Raizer (7) define the width of a shock wave as the distance at which the viscosity and heat conduction become negligible. This occurs within a few molecular collisions in a gas. The internal modes of gaseous explosive molecules are also becoming excited: translational modes (a few collisions); rotational modes (tens of collisions); and vibrational modes (hundreds of collisions). These equilibration processes have long been studied in shock tubes (8). Internal energy equilibration is now being studied in shocked liquid and solid explosives by Dlott et al. (9) and Fayer et al.(10). In condensed phases, the phonon modes are excited followed by multi-phonon excitation of the lowest frequency vibrational modes and then the higher frequency modes by multi-phonon up-pumping and internal vibrational energy redistribution (IVR)(11). Once the explosive molecules have attained vibrational equilibrium, chemical decomposition can begin.

For gaseous explosives, these equilibration processes which precede chemical reaction are easily measured since they can be lengthened to nanosecond or even microsecond time frames by dilution with inert gases or by the use of low initial pressures. The calculation of these states is also straight forward, because the perfect gas law applies. The initial reaction rates for the dissociation of the weakest chemical bond present in the explosive molecule/mixture are also easily measured in shock tube experiments and calculated using unimolecular Arrhenius chemical kinetics. If the shock wave heats the explosive molecules to temperatures at which sufficient dissociation occurs before the shock compression ends and rarefaction cooling begins, the newly formed atoms react with surrounding molecules. An exothermic chain reaction process follows in which reaction product gases are formed in highly vibrationally excited states (12). These excited products either undergo reactive collisions with the surrounding explosive molecules or non-reactive collisions with their neighbors in which one or more quanta of vibrational energy is transferred. Some collisions are "super-collisions"(13) in which several quanta of vibrational energy are transferred. Since reaction rates increase rapidly with each quanta of vibrational energy available, reactive collisions dominate and the main chemical reactions are extremely fast. Once the chain reaction process is completed, the remainder of the reaction zone is dominated by the de-excitation of highly vibrationally excited product molecules as chemical

equilibrium is approached. This de-excitation process controls the length of the reaction zone and provides the chemical energy necessary for shock wave amplification during shock-to-detonation transition (SDT) and self-sustaining detonation.

The Non-Equilibrium Zeldovich - von Neumann-Doring (NEZND) theory of detonation (12,14-17) was developed to explain the various non-equilibrium processes that precede and follow chemical energy release in self-sustaining detonation waves. As pressure wavelets pass through the subsonic reaction zone, they are amplified by vibrational de-excitation processes. The opposite effect – shock wave damping by a non-equilibrium gas that lacks vibrational energy after expansion through a nozzle – is a well-known phenomenon (7). The pressure wavelets then interact with the main shock front and replace the energy lost during compression, acceleration and heating of the explosive molecules. During shock initiation, this interaction process increases the shock front pressure and velocity. If the initial shock wave is accelerated to a velocity at which chemical reaction occurs close to the front, then self-sustaining detonation occurs. The pressure wavelet amplification process then provides the required chemical energy by developing a three-dimensional Mach stem shock front structure. This leading shock wave front is still a “wake-up call” or “trigger” for explosive reaction, but it is also a “burden” for the explosive molecules to sustain at a constant supersonic velocity with their energy.

The three-dimensional structures of detonation waves have been observed for gaseous, liquid and solid explosives (18). In gaseous detonations, the details are very well known and several excellent reviews of the subject are available (19). For liquid and perfect single crystal solid explosives, the situation is much more complex and thus more difficult to observe and calculate than in gases. The high initial densities of the condensed phases make the measurement and calculation of the states attained behind a shock wave more difficult, because the processes now take tens and hundreds of picoseconds and the perfect gas law does not apply. The distribution of the shock compression energy between the potential (cold compression) energy of the unreacted liquid or solid and its thermal energy is a complex function of shock strength. The lack of voids, cracks, particle boundaries, etc. eliminates “hot spot” formation as an initiation mechanism. If the shock compressed state lasts long enough for exothermic reaction to begin at this shock

temperature, initiation of occurs at or near the boundary of the explosive charge in the molecules first impacted by the shock. This “thermal explosion” creates a “superdetonation” wave that propagates through the precompressed explosive at a velocity in excess of its equilibrium Chapman-Jouguet (C-J) velocity. When this wave overtakes the leading shock wave, its velocity decreases rapidly until steady state velocity is attained. This phenomenon has been measured and calculated for several detonating liquids (20) and solid perfect crystals (21). Liquid explosives exhibit a wide range of shock sensitivity (22). Perfect single crystals of relatively sensitive solid explosives like PETN can be shock initiated (21), but single crystals of HMX can not be initiated by a detonation wave from an HMX-based plastic bonded explosive (6).

The “induction” time for the initial “thermal explosion” can be calculated using the high pressure, high temperature transition state theory. Experimental data for unimolecular gas phase reaction rates under low temperature shock conditions obeys the usual Arrhenius law:

$$K = A e^{-E/RT} \quad (1)$$

where K is the reaction rate constant, A is a frequency factor, E is the activation energy, and T is temperature, at low temperatures, but “fall-off” to less rapid rates of increase at high temperatures (23). Nanosecond reaction zone measurements for solid explosives overdriven to pressures and temperatures exceeding those attained in self-sustaining detonation waves have shown that the reaction rates increase very slowly with shock temperature (24). Eyring (25) attributed this “falloff” in unimolecular rates at the extreme temperature and density states attained in shock and detonation waves to the close proximity of vibrational states, which causes the high frequency mode that becomes the transition state to rapidly equilibrate with the surrounding modes by IVR. These modes form a “pool” of vibrational energy in which the energy required for decomposition is shared. Any large quantity of vibrational energy that a specific mode receives from an excitation process is shared among the modes before reaction occurs. Conversely, sufficient vibrational energy from the entire pool of oscillators is statistically present in the transition state long enough to cause reaction. When the total energy in the vibrational modes equals the activation energy, the reaction rate constant K is:

$$K = (kT/h) e^{-s} \sum_{i=0}^{s-1} (E/RT)^i e^{-E/RT} / i! \quad (2)$$

where  $k$ ,  $h$ , and  $R$  are Boltzmann's, Planck's, and the gas constant, respectively, and  $s$  is the number of neighboring vibrational modes interacting with the transition state. The main effect of this rapid IVR among  $s+1$  modes at high densities and temperatures is to decrease the rate constant dependence on temperature. Reasonable reaction rate constants were calculated for detonating solids and liquids using Eq. (2) with realistic equations of state and values of  $s$  (15). For the lower temperatures attained in shock initiation of homogeneous liquid and solid explosives, the reaction rate constants calculated using Eq. (2) are larger than those predicted by Eq. (1). Reaction rate constants from Eqs. (1) and (2) are compared to induction time results for gaseous norbornene, liquid nitromethane, and single crystal PETN in Figs. 1 – 3, respectively (16). Despite uncertainties in the calculated shock temperatures for various equations of state, it is clear that Eq. (2) agrees quite well with all three sets of data using reasonable values of  $s$ . Thus high pressure, high temperature transition state theory accurately calculates induction times for shock induced reactions during shock initiation and detonation of homogeneous gaseous, liquid, and solid explosives.

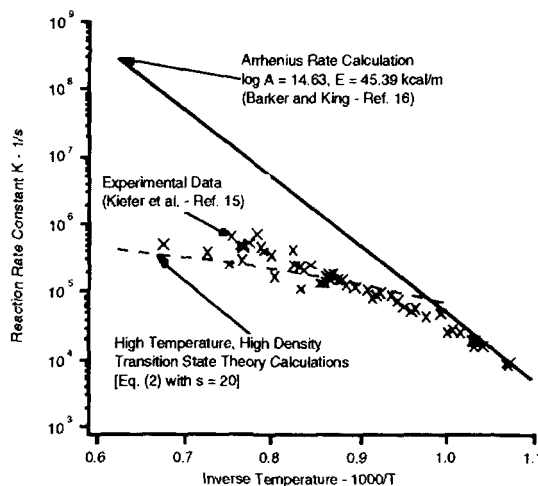


FIGURE 1. Reaction rate constant versus inverse temperature for the unimolecular decomposition of norbornene

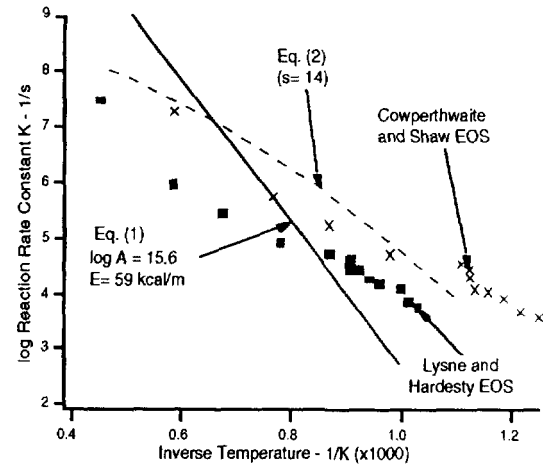


FIGURE 2. Reaction rate constants for nitromethane as functions of shock temperature

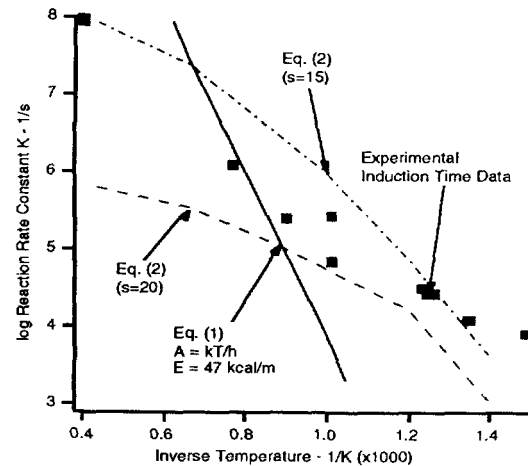


FIGURE 3. Reaction rate constants for single crystal PETN as functions of shock temperature

## HETEROGENEOUS EXPLOSIVES

For heterogeneous explosives (liquids with bubbles or suspended solid particles and pressed or cast solids with voids, binders, metal particles, etc.), an initiating shock wave does not have to heat the entire material to the point of thermal explosion. Thermal energy is concentrated in local sites by the physical processes of void collapse, friction, shear, dislocation pile-up, etc. Liquid explosives which contain bubbles can undergo partial reactions known

as low velocity detonation (LVD) at heated sites created by collapsing voids. LVD can propagate long distances in pipes and is a major safety concern. LVD can cease to propagate or transition to full detonation in various scenarios (26).

It has long been known that shock initiation of solid explosives is controlled by ignition of hot spots (3). How large and how hot does a hot spot have to be to react and begin to grow? Critical conditions for the growth or failure of hot spots in HMX- and TATB-based explosives have been calculated using multistep Arrhenius kinetic chemical decomposition models derived from thermal explosion experiments (27). Figure 4 shows the calculated critical spherical hot spot temperatures in HMX and TATB. Once ignited, the growth rates of reacting hot spots into neighboring solid explosive particles and the interactions of several growing hot spots have been calculated for various geometries (28). Figure 5 shows the times required for spherical HMX particles of various radii to complete deflagrate inwardly under various boundary temperature conditions. These relatively long times show that large explosive particles must fragment, producing smaller particles with more reactive surface area for hot gaseous reaction products to ignite. As growing hot spots coalesce at high pressures and temperatures, the transition from shock induced reaction to detonation occurs very rapidly. The buildup of pressure and particle velocity behind the shock wave front during shock initiation has been thoroughly studied using embedded gauge (29,30) and

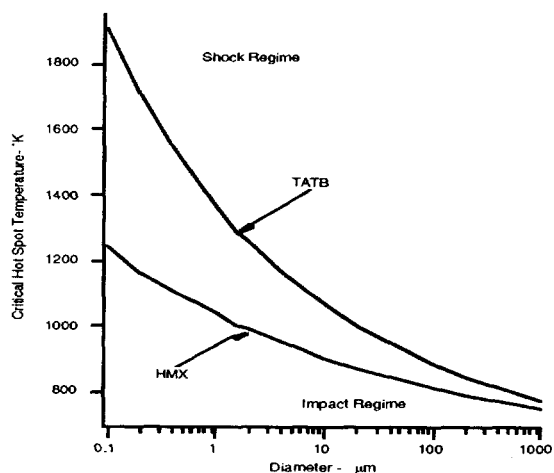


Figure 4. Critical spherical hot spot temperatures in HMX and TATB at various diameters

laser interferometric (31) techniques. These reactive flows have been modeled in multidimensional codes by the Ignition and Growth model of shock initiation and detonation (32). Figure 6 shows measured and calculated pressure histories obtained for a shock initiation experiment on HMX-based LX-04 (29).

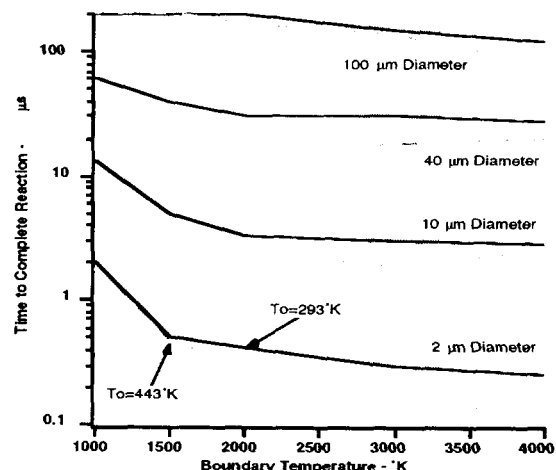


FIGURE 5. Reaction times for HMX particles

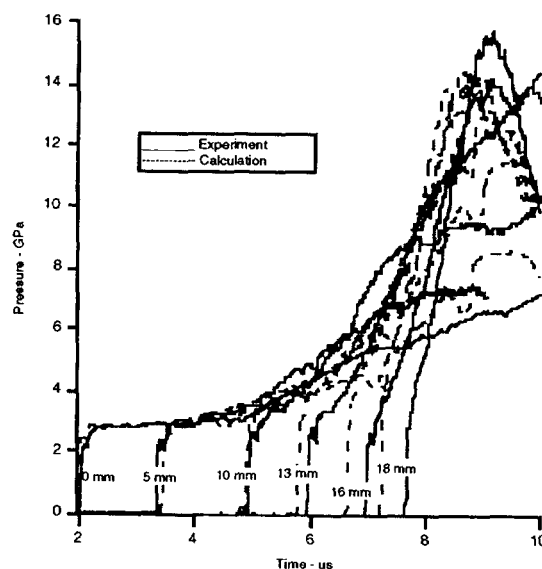


FIGURE 6. Pressure histories for ambient temperature LX-04 shock initiated by a Teflon flyer plate at 0.956 mm/μs

Detonation wave reaction zone structures in solid explosives and their metal acceleration properties have also been measured by embedded gauges and

laser interferometry and calculated by the Ignition and Growth model (33). Figure 7 shows the measured and calculated interface velocity histories for detonating LX-17, a TATB-based explosive, impacting various salt crystals (33). Figure 8 illustrates the measured and calculated free surface velocities of 0.267 mm thick tantalum discs driven by 19.871 mm of detonating LX-17

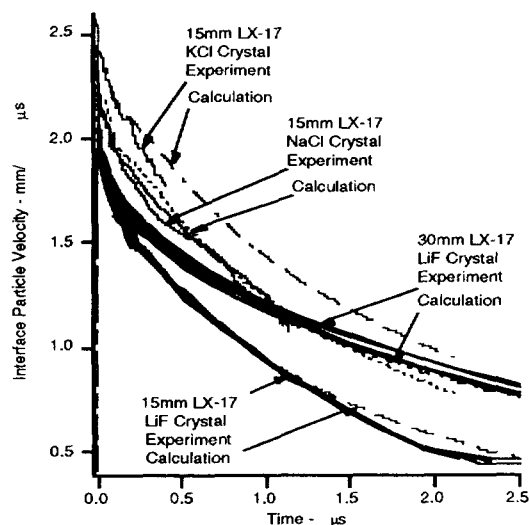


FIGURE 7. Interface particle velocity histories for detonating LX-17 and various salt crystals

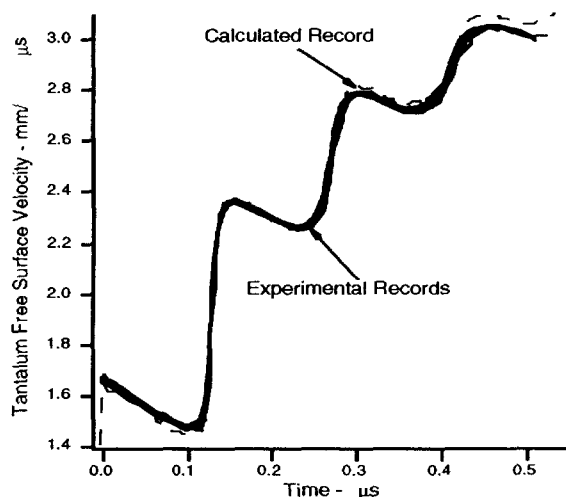


FIGURE 8. Free surface velocities for 0.267 mm thick tantalum discs driven by 19.871 mm of LX-17

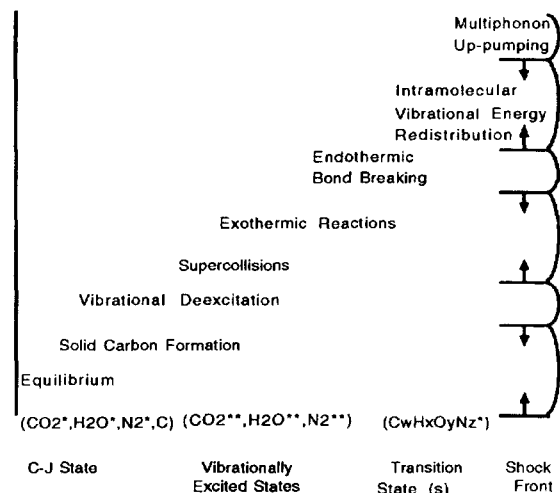


FIGURE 9. The Non-Equilibrium Zeldovich - von Neumann-Doring (NEZND) model of detonation for condensed phase explosives

Since the main application of detonating solid explosives is to accelerate metals and other materials to high velocities, an understanding of the unreacted shock state (the "von Neumann spike"), the pressure profile in the chemical reaction zone, and the subsequent expansion of the reaction products as they deliver their momentum to the metal is essential. Currently the one-dimensional averages of these properties are known to within a few percent with several nanosecond resolution (21,31,33,34).

Due to solid particle interactions, one expects the detonation front structure to be more complex and less regular in heterogeneous explosives than in homogeneous ones. The sub-nanosecond techniques needed to resolve this wave structure are becoming available. Figure 9 illustrates the various processes that precede and follow exothermic chemical reactions behind each wavelet of the three-dimensional structure comprising the reaction zone of a condensed phase detonation wave. Eventually all of these non-equilibrium physical and chemical mechanisms, along with those that have not been identified as yet, will need to be measured experimentally and modeled in advanced multidimensional reaction flow models. Then the interactions of shock waves with explosive molecules and vice versa can be better understood. This understanding may lead to the production of safer, more energetic explosive molecules and formulations.



## FUTURE RESEARCH

While a great deal has been learned in recent years about the interaction of shock waves with explosive molecules, greater spatial and time resolution is needed in shock wave experiments and calculations. For understanding low velocity impact ignition mechanisms, the relative roles of void collapse, friction, shear, dislocation pile-up, etc. need to be determined by clever experimentation. Many of these postulated hot spot formation mechanisms depend upon the magnitude of the viscosity in and behind shock wave fronts, which has not yet been measured for shock waves in condensed phase explosives. If the dominant hot spot mechanism (or mechanisms) can be identified experimentally and successfully modeled, modifications to existing explosive formulations can be made. New processes and new materials (explosives, binders, additives, etc.) can be developed to produce safer products.

Since chemical reaction rates are controlled by the local temperature of a region of molecules, the most important need is for experimental measurements of temperature in all regions of shocked explosives: in and around hot spots; in deflagration waves; in the reactive flows behind shock fronts and in detonation waves. With this type of data, improved equations of state and all-Arrhenius reactive flow models can be developed to better predict the effects of shock waves on explosive molecules and vice versa (28). Eventually it will become possible to model shock induced reactions as thermal decomposition mechanisms are modeled today by identifying intermediate reaction product species and following their concentration changes. To do this effectively, nanosecond or faster time resolved experimental data on the rates of consumption of the unreacted explosive, the concentrations of intermediate species, and the rates of production of the final stable products is needed. Accurate determination of the three-dimensional structures of detonation waves in condensed phase explosives is required to determine how much detail must be included in reactive flow models to yield more realistic and predictive simulations.

## SUMMARY

This short review can only begin to address the complex question: What is a shock wave to an explosive molecule? Through several compression and heating mechanisms, a shock wave is the "wake-

up call" or the "trigger" by which the exothermic power of the metastable explosive molecule is unleashed. Many possible outcomes of the initial hot spot formation process are possible: no reaction; shock desensitization; a weak explosion; a violent explosion; deflagration; shock wave formation or amplification; and transition to detonation. Detonation is of course the desired result of an intentional shock initiation but must be avoided at all costs during unintentional initiations (accidents). At the maximum rate of energy delivery in a detonation wave, the leading shock wave initiates the chemical reaction but then must be sustained by the chemical energy released. This chemical energy is initially released into highly vibrationally excited reaction products, whose relaxation to chemical equilibrium must amplify pressure wavelets propagating through the subsonic reaction zone. Understanding this intimate connection between non-equilibrium chemical kinetics and the three-dimensional detonation wave structure is the key to developing improved reactive flow models and safer, more powerful explosives.

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